

Comment on “Atomic jumps in quasiperiodic $\text{Al}_{72.6}\text{Ni}_{10.5}\text{Co}_{16.9}$ and related crystalline material”

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We disagree with a number of statements by Dolinšek et al. about the specificity of phason dynamics in quasicrystals (QCs).

We are surprised by the lack of foundation and clarity of some formulations in a recent paper by Dolinšek et al. [1] about phason dynamics in QC. It must be clear that there is no ground for an allegation that the dynamical signal observed in references [2] would be due to vacancy diffusion rather than phason hopping. In fact, the Q -dependence of the neutron-scattering signal indicates that the atomic motion remains confined in space, while its temperature dependence is unusual and not typical of vacancy diffusion. Furthermore, the diffusion constants that one experimentally observes are much lower than the ones one should reasonably expect on the basis of the observed hopping rates if this hopping were due to vacancy diffusion. Indeed, the hopping is exceptionally fast, while the observed diffusion constants just take values that could be qualified as standard for metallic compounds. The authors are stating this themselves, such that they do not seem to question all these matters. It is therefore puzzling what the statement about a remarkable similarity between the activation energy of the hopping process and the enthalpy for the formation of a vacancy in pure Al is supposed to imply. What is the aim of this statement about what probably is merely a numerical coincidence? We must mention that the numbers quoted from our work are *not* the *activation* energies of the hopping process, but *assistance* energies. Moreover, the value of 0.6 eV quoted for AlCuFe is related with Cu rather than with Al hopping, and the assistance energies we reported for AlPdMn are not remarkably close to 0.6 eV. On the same footing it is not clear what the statement that high temperature phason hopping would “interfere” with other processes is supposed to imply.

The authors claim that it has been shown that d-AlCoNi contains a large amount of “vacant sites”. The “vacant sites” in question are just phason sites, [3] such that this claim contradicts the earlier introductory statement of the authors that no empty lattice site is involved in the concept of a phason flip. That we are dealing here with phason sites rather than with vacancies is not an issue and leaves no space for any confusion of the kind that would seem to emanate from the presentation of the authors. The confusion is produced by the undifferentiated terminology “vacant site” which the authors use for both phason sites and vacancies.

Other poorly justified statements have been made by these authors to promote the idea that QCs would contain a large amount of vacancies. [4,5] This shows that

this statement has not been deduced by unbiased logical deduction from scientific observation, but that it is a preconceived postulate the authors try to validate. This postulate has a pivotal function in the argument of the authors. It must serve to lend credibility to an analogy they want to impose between B2-based crystals (which might contain up to 12 percent structural vacancies) and QCs (where nothing of that order of magnitude has ever been established). [6] This then should permit to incorporate phason dynamics into a much broader class of trite hopping phenomena.

The authors argue that in contrast with high-temperature data, the low-temperature data would not be subject to “interference”. This cannot conceal the lack of conclusive prove for the attribution of the low-temperature NMR signal to phason dynamics, as reflected in the caution of the statement that the data are *compatible* with phason dynamics. NMR data do not provide much information about characteristic distances as they do not yield Q -dependent information. The time scales accessed are quite remote from the time scales that can be accessed by other techniques such that cross-checking or use of complementary information to validate the claims is not possible. This is unfortunate as the existence of such slow low-temperature dynamics is certainly intriguing. The only thing we really know with certainty is that there is some slow (local) relaxation with a low activation energy. E.g. the idea of a small local shift of the Al atom in response to a slowly fluctuating or diffusing strain field that includes its environment is equally compatible with their data. One can think of other phenomena that could be compatible with the data. Without wanting to reinterpret the NMR data accordingly, we may mention that one type or another of slow dynamics remaining unfrozen at low temperatures is observed in many systems, including QCs, e.g. in the form of tunneling states. [7] Above a few degrees K the coherent tunnelling may cross over to a thermally activated process. The low temperature region is thus not as exempt of the possibility of “interfering” dynamics as the authors suggest. It has still not been proved experimentally that tunneling states in QCs are not phasons. This illustrates the difficulty of making assignments. In glasses tunneling states are conceived as small simultaneous shifts in the positions of groups of atoms bringing about a transition between slightly degenerate con-

figurations. The precise detailed geometrical picture of the motions involved in such processes remains unknown. This kind of lack of information is a recurrent theme in slow dynamics and due to the physical limitations of the available experimental techniques. From all this we may retain a neglected possibility of explanations in terms of small, non-phasonic atomic shifts, whose amplitudes explore a continuum rather than a discrete set.

The suggested uniqueness of interpretation of the NMR data is also contradicted by other NMR data of the authors. In fact, in reference [8] they observed a temperature behaviour of an NMR signal that was not compatible with their interpretation of it in terms of phason dynamics. The AlPdMn phase can be magnetic, a fact that offers seeds for an alternative explanation. [9] In a subsequent paper they stated [10] that this scenario could not be proved, and concluded that the origin of the signal was not understood, but probably due to unusual magnetic properties of Mn atoms. They also stated that the data from AlPdRe are of a different origin than those from AlPdMn, despite possible similarities in the time scales. This illustrates that other phenomena that are not frozen can exist at low temperatures and wrongfoot the interpretation of the data. It is not because two signals look similar that they cover similar physics: What is more similar to a relaxation time with an activation energy than another relaxation time with an activation energy? But in many experimental techniques, relaxation times and activation energies are all the hard data resume to. Now the authors have established the existence of similar signals in B2-based AlNiCu, which despite all possible claims definitely does not support phason dynamics. If following them we discard interference from vacancy motion, we are faced with the problem of elucidating the origin of an NMR signal in the B2-based phase that is not due to phasons nor to vacancy hopping. The fact that such unclearly identified atomic dynamics can give rise to a signal that is similar to the one observed in QCs is (in view of the unappropriateness of the comparison) not a proof that the assignment of the authors is wrong, but does reveal that its uniqueness is not sufficiently substantiated. We may note that the theory of Jarić and Nelson for diffuse scattering [11] is based on the ansatz that phason dynamics is frozen. Hence the assignment of the authors also summons for a rethinking of matters that have been directly linked to the validity of the random tiling model.

In summary, the main conclusion of the authors that phason dynamics is not QC-specific cannot be reached on the basis of the data presented. But even if such a conclusion could be reached, nobody would understand QC-specificity in the singular, restrictive sense the authors want to give to it. Their concern of uniqueness is much less in order than the one about the assignment of the NMR signal. In fact, phasons correspond to atomic jumps in double-well potentials, whose minima are separated by a distance shorter than the interatomic distances. Nobody claims that such double-well potentials

giving rise to atomic jumps would be a rarity in solid state physics. The example of the hydrogen bond has been well known for a long time. One of the original features of QCs is that the presence of these short-distance double wells is an integrated part of the quasilattice. Conceptually, a simple B2-based lattice with vacancies does not imply the existence of short-distance double wells. It would thus appear that examples of signals from B2-based phases do not capture the essence of the non-uniqueness of the dynamics of such double-well potentials. As already stated, there is more to the similarity of the physics than a similarity of relaxation times.

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